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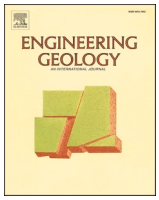

Ageing/thixotropy effects on strength and compressibility in Leda clay and polymer-amended clayey tailings

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ABSTRACT

Evidence of ageing has been reported in natural deposits of sedimentary clays, dredged sediments, and in clay-rich oil sands tailings, and such time-dependent behaviour has been observed to influence their long-term settlement and strength development characteristics. The ageing phenomenon can, therefore, have important implications for the reclamation of such deposits. This research investigated ageing in polymer-amended tailings and Leda clay monitoring relatively small (10 cm high) laboratory samples for over 100 days. Both tailings and natural clay were found to develop increased strength, sensitivity, and higher pre-consolidation pressure and reduced compressibility. The correlation between fall cone shear strengths and pre-consolidation pressures was strong for both the natural clay and the clayey tailings, though this ratio was different in the tailings and the clay. The Leda clay appeared to reabsorb some of its bleed water, which was found to be due to the transition of some free water to absorbed water after 80 days. Using a concept developed from ageing experiments in artificial clays, it was found that shear strengths measured after an "early equilibrium state" correlated strongly with shear strengths measured near the end of ageing. This is promising for quantifying the potential for ageing on compressibility without having to resort to year-long experiments.

1. Introduction

Slow consolidation of tailings or dredged sediments poses large challenges for the management of impoundments, both in terms of how the capacity of such impoundments changes over time, and challenges associated with reclamation work, which requires sufficient strength for trafficability and long-term stability. Although the void ratio-effective stress relationship and hydraulic conductivity-void ratio relationship are the two most important properties that govern the consolidation behaviour of such materials, creep and thixotropic behaviour of tailings have also been found to influence the consolidation performance of tailings deposits (Jeeravipoolvarn, 2005; Jeeravipoolvarn et al., 2009; Miller, 2010). Several models for creep-consolidation exist in the literature (Yin and Graham, 1994; Vermeer and Neher, 1999; Hinchberger and Rowe, 2005); these models reproduce creep effects by implicitly or explicitly modifying the apparent compressibility curve as a function of strain rate. Ageing, or the increase in pre-consolidation pressure over time, is mostly studied in the soil mechanics literature with reference to differences in compressibility and strength between the intact and reconstituted states of natural clays (Locat and Lefebvre, 1986; Leroueil and Vaughan, 1990; Burland, 1990; Leroueil et al., 1996; Liu and Carter, 1999; Sorensen, 2006), which bears practically on appropriate laboratory procedures to evaluate settlement and strength in those deposits. Rate of ageing is less important to natural clays; however, freshly deposited tailings, dredged sediments, or similar materials are by their very nature "young" materials, and therefore information on their ageing may be required to correctly estimate their long-term consolidation behaviour.

In this paper, ageing has been investigated in terms of changes in the compressibility behaviour of a material independent of density. Changes in the compression curve over time have been studied by Zeng et al. (2016) for dredged sediments, and by Salam et al. (2018a, 2018b), Jeeravipoolvarn (2005), Jeeravipoolvarn et al. (2009), and Miller (2010) for different kinds of tailings. Similar phenomena are thought to contribute to ageing and thixotropy, where the latter is usually defined as the recovery of undrained shear strength after remoulding under constant moisture, constant volume, and constant temperature conditions, due to time-dependent reorganization of the fine clay particles because of the electrochemical forces acting between the particles in a clay-water system (Mitchell, 1960). Skempton and Northey (1952)

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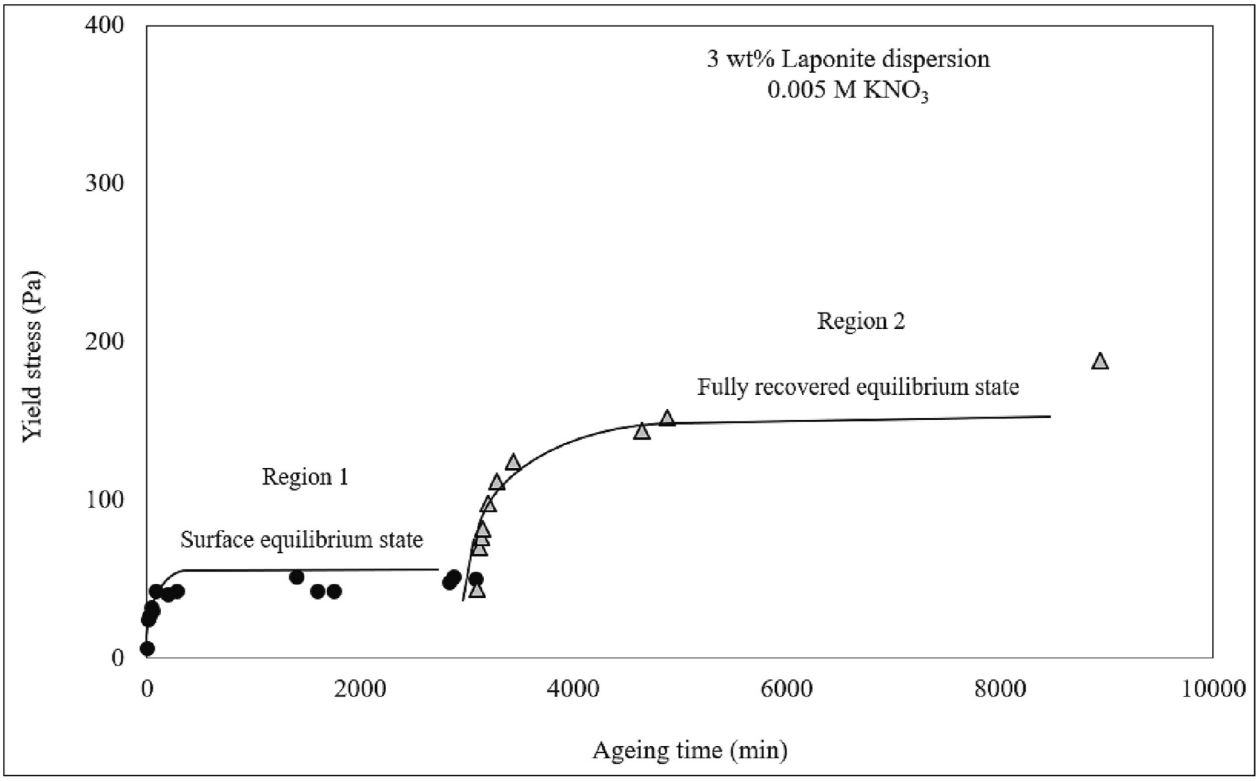

Fig. 1. Laponite dispersions showed an ageing behaviour, which was characterized by a strength development in the sample in two distinct phases (adapted from Au and Leong, 2015).

Table 1
The properties of unamended (amended) FFT and Leda clay samples.

Parameters	Unamended (amended) FFT	Leda clay
Initial solids content (%)	31	57
Initial water content (%)	225	77
Specific gravity	2.12	2.7
Liquid limit (%)	60 (70–75)	51
Plastic limit (%)	28 (35–40)	28
Plasticity index (%)	32	23
Clay content (%)	28	71
D_{90}, D_{60}, D_{50}, D_{10} (μm)	100, 11, 6, 0.8	10, 1, <1, <1
Clay mineralogy	Illite (28%), Kaolinite (72%)	Illite (83%), Kaolinite (11%)

attributed the thixotropic effect in the soil to the gradual rearrangement of the particles under the influence of bonding forces such as van der Waals forces. Recent studies on thixotropic ageing in reconstituted clays also showed that thixotropic ageing was associated with particle reorientation and the stiffness of the samples increased over time because of the decrease in inter-particle distance and the subsequent development of bonding between particles (Anik, 2019; Alam et al., 2021). Thixotropic strength gain over time has also been reported in unamended tailings (Jeeravipoolvarn, 2005; Jeeravipoolvarn et al., 2009; Miller, 2010), and in dredged clays (Zeng et al., 2016; Shahriar et al., 2018; Shahriar and Jadid, 2018). The magnitude of thixotropy is known to increase with decreasing water content in tailings (Banas, 1991; Suthaker, 1995; Suthaker and Scott, 1997) and in soils (Skempton and Northey, 1952; Seng and Tanaka, 2012), down to the liquid limit, but then subsequently decreases.

Generally, the rate of ageing or strength/stiffness development in clays has been observed to decrease over time (Skempton and Northey, 1952). Interestingly, outside of the soil mechanics literature, ideal suspensions of artificial clays such as Laponite are studied in great depth to understand ageing behaviour in colloidal suspensions (Abou et al., 2001; Ruzicka et al., 2007; Pujala and Bohidar, 2013; Au and Leong, 2015). Ageing is understood to progress from a meta-stable state, that may be induced either by shearing a colloidal suspension or by rapidly changing its concentration or temperature. Ageing occurs in such systems through either the re-ordering of particles, which are influenced by electrochemical forces, and may form different types of structures, from large flocs to sample spanning networks; or from increases in the strength of existing contacts (Bonacci et al., 2020). Ageing may in part progress by Brownian motion (Abou et al., 2001; Pujala and Bohidar, 2013; Weeks, 2017). Ageing may occur in either repulsion dominated systems (termed glasses), or attraction dominated suspensions (gels). Generally, the rate of ageing, and the increase in strength, increases with the increasing magnitude of net inter-particle electrical chemical forces (Tanaka et al., 2005; Mewis and Wagner, 2012) and with increasing concentration of solids (decreasing void ratio or water content) (Abou et al., 2001; Pujala and Bohidar, 2013; Au and Leong, 2015).

It has been observed in ageing experiments on ideal clay suspensions with Laponite that the strength development in this type of experiments exhibits two modes: An initial increase in yield stress or shear strength or small-strain elastic modulus which plateaus, followed by a subsequent increase in strength or modulus, which eventually also plateaus (Fig. 1, adapted from Au and Leong, 2015). The first equilibrium state sometimes called the “Surface Chemical equilibrium (SCE)” state, is attributed to the time required for the saturation of clay surface by ions after initial sample preparation (Goh et al., 2011; Au and Leong, 2015).

The goal of the present paper is to understand ageing in polymer-amended clayey tailings. For example, it is the state of practice of tailings deposition in Canada’s oil sands industry to mix polymers and other amendments into the clayey fraction of tailings to improve tailings consolidation. However, as future deposits of these treated tailings are envisaged to be quite deep (> 100 m), the bearing of time-dependent

(a)

Measurements taken before breaking columns	Tests conducted after breaking columns
▪ Height of the sample in the column ▪ Weight of the sample in the column ▪ Height of the water-FFT interface	1. Total column: ▪ Water/solids content ▪ Net water release ▪ Solids settlement ▪ Void ratio 2. For each 1 cm layer: ▪ Water/solids content 3. Topmost 1 cm layer: ▪ Scanning electron microscope (SEM) ▪ Fall cone test 4. Bottommost 1 cm layer: ▪ Oedometer test

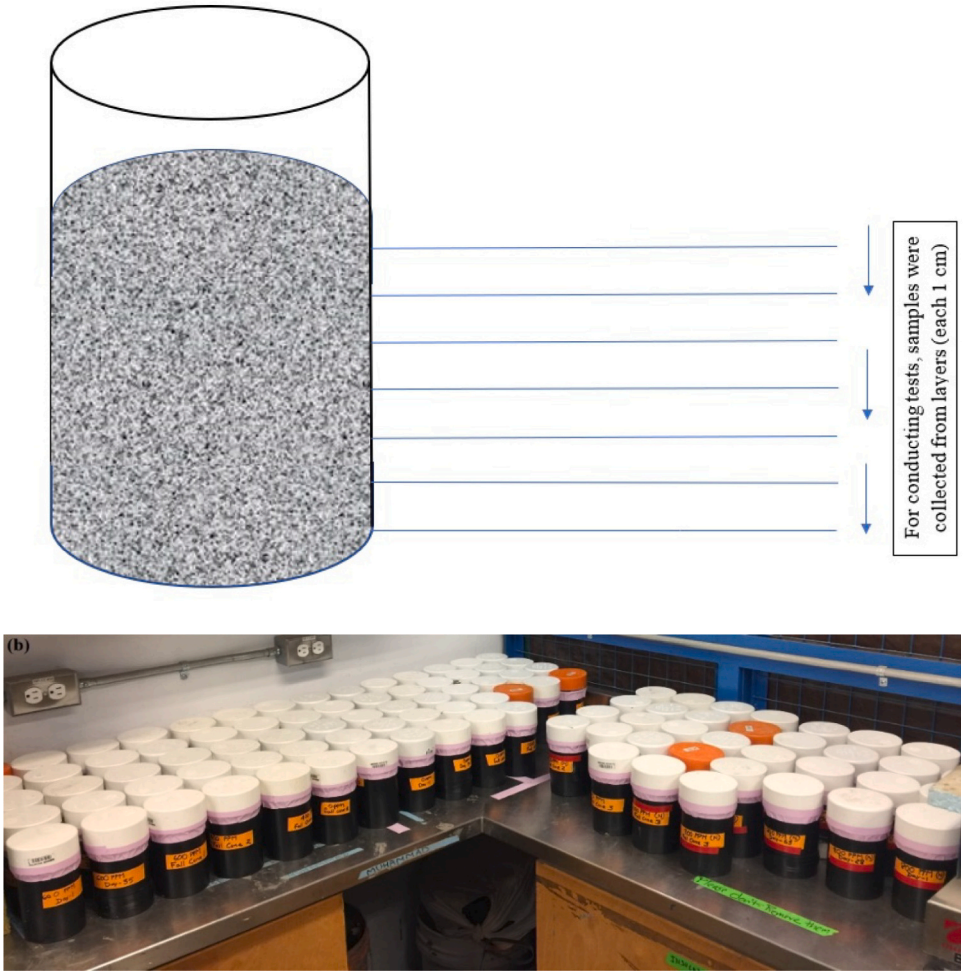

Fig. 2. (a) A schematic diagram of the polypropylene column used for destructive testing; (b) all the replicates (both FFT and Leda clay).

effects such as ageing may be very important. Hence this study examines whether the ageing is affected by varying polymer dosages. Leda clay is also studied in the experimental matrix. Leda clay deposits are known to exhibit ageing in their natural undisturbed state (Quigley and Thompson, 1966; Yong and Nagaraj, 1977) and have similar geotechnical properties and mineralogy to the clayey tailings. Readers should be aware that Leda clay is now often designated as 'Champlain sea clay' in some of the more recent literature (e.g., Liu et al., 2021; Monsif et al., 2021), though much of the classic literature refers to this clay as 'Leda clay' (Quigley and Thompson, 1966; Crawford, 1968; Walker and Raymond, 1968; Yong and Nagaraj, 1977). The name 'Leda clay' has been used in this paper for convenience.

The principal tests reported in this paper are water content depth profiles, fall cone shear strength, and compressibility curves from oedometer tests obtained from the 10 cm columns. These columns for all soil and tailing types were allowed to age over 100 days, and up to 650 days for some specific samples. The materials and the methods used for the tests have been described in detail in the following sections (i.e., Section 2 and Section 3 respectively). In the Results section (i.e., Section 4), after presenting how the dewatering and settlement, undrained shear

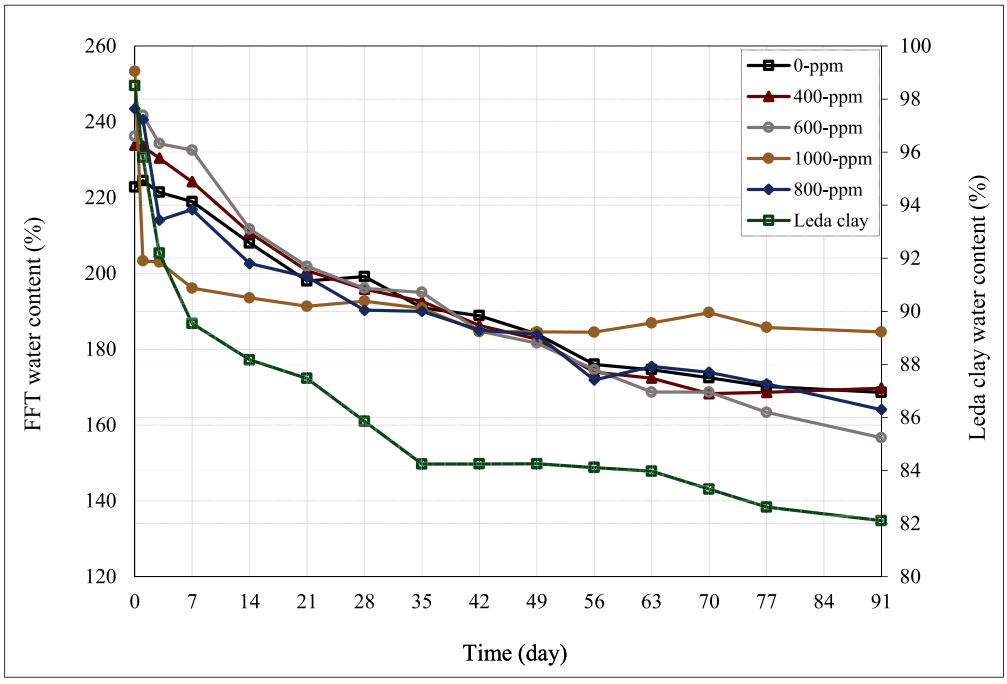

Fig. 3. Water contents in the Leda clay and fFFT 10 cm tall columns.

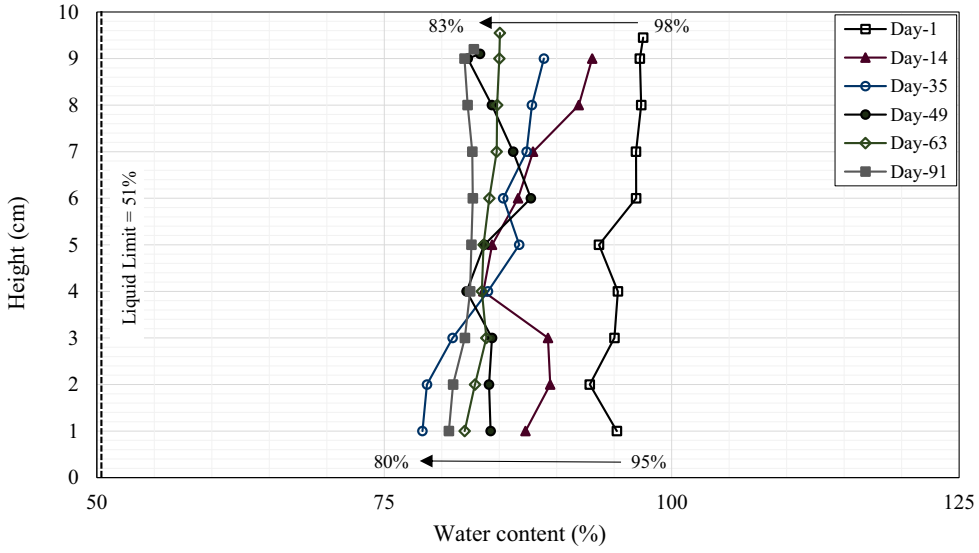

Fig. 4. Water content depth profiles in Leda clay samples.

strength, and consolidation behaviour changed over time differently depending on material type and polymer dosage, the criteria for determining "early equilibrium state" (EES) strengths from individual strength curves were defined. In the Discussion section (i.e., Section 5), a logistic function was proposed as a descriptive equation to determine the EES strength for ageing data sets where EES can not be obtained using those criteria. The shear strength data of both the original and the literature data is then normalized with the EES and compared. Finally, the influence of the magnitude of the change in the compressibility curves on the consolidation of tailings deposits is briefly discussed.

2. Materials

2.1. Oil sands tailings and Leda clay

Oil sands tailings samples were collected from a tailings pond in Northern Alberta, Canada, and shipped to Carleton University in Ottawa, Ontario, Canada. The Leda clay used in the study was collected from the Navan landfill in Ottawa, Ontario, Canada. Samples were sent to an external laboratory for conducting different characterization tests including pore-water chemistry, X-ray diffraction analysis, Methylene blue index test, and Dean-Stark analysis. The properties of unamended fluid fine tailings (FFT) and Leda clay samples are summarized in Table 1.

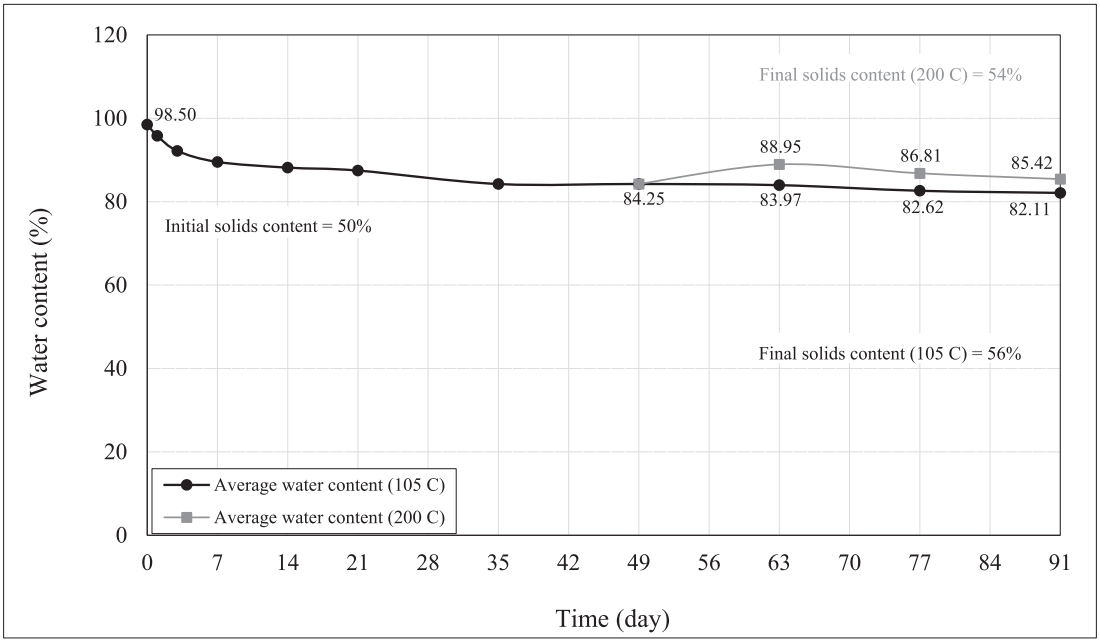

Fig. 5. The changes in water content in the Leda clay samples (measured using 105 °C, 200 °C).

2.2. Polymer stock solution and polymer mixing methods

An anionic polyamide-based polymer (A3338) was used to prepare the polymer-amended FFT samples using variants of standard industry protocols to replicate field mixing conditions (Mizani et al., 2017). A polymer stock solution (0.4% concentration) was prepared by adding polymer to the deionized water and stirring the solution using a jar tester for an hour. 2 different mixing methods were used to mix the polymer solution with the FFT. The first mixing method (Method#1) involved mixing the FFT with the required amount of polymer solution at 250 rpm for 20 s. The second method (Method#2) involved mixing at 2 different steps: (a) 320 rpm for the first 10 s and (b) 20 rpm for the next 10 s. 5 different dosages of polymer, including 0-ppm (unamended FFT, which was used as a control sample) were used for this experiment. The differences due to mixing protocols were not detectable in the results (Salam, 2020).

3. Methods

Samples were prepared by mixing of polymer solutions into the tailings at their existing water content by the methods described in the previous section, or by increasing the water content of the Leda clay to twice its liquid limit (LL) by mixing the required amount of deionized water into the clay by hand mixing for 15 min. Samples were then placed into 10 cm tall 7.5 cm internal diameter polypropylene columns using a single-drainage boundary condition. Polypropylene is chosen as it exhibits minimal adherence between clay materials and the surface compared to the materials such as acrylic (Salam, 2020). A relatively short height of 10 cm was selected for the deposited material because 10 cm allows for relatively rapid self-weight consolidation, though providing sufficient sample to provide good data post-destructive testing. Such short column height still allowed for the construction of water content/density profiles with depth upon destructive testing. Other consolidation studies using thicker columns (up to 0.50 cm) have been conducted; these however take considerable time to complete primary consolidation, often longer than 2 months for polymer-amended FFT.

There were equal numbers of columns (14) for each polymer dosage and the Leda clay samples. The columns were kept covered at the top with the help of a tight lid. The space between the lid and the surface of the deposited clays/tailings was also kept closed to prevent the escape of water vapour. The non-porous and sealed bottom in the single-drainage columns prevented downward drainage. The mass of each column was recorded daily during the first two weeks and then weekly, showing negligible changes in mass even over 100 days; for example, in the 600-ppm sample, only 3.8 g were lost from a total sample weight of 533 g after 91 days. Every week one column from each dosage was selected for destructive testing and samples were collected from each 1 cm layer of FFT for conducting different tests including oedometer tests, fall cone tests, and low vacuum scanning electron microscopy (Fig. 2). The lines in Fig. 2(a) referred to those 1 cm layers of flocculated FFT (fFFT). Water contents were measured to different temperatures (105 °C, 200 °C, and 550 °C) to see if changes in the water phase were occurring.

Self-weight consolidation was monitored in the selected samples using a pore-water pressure sensor embedded in a subset of the 10 cm columns. Pore-water pressure sensors were located at an elevation of 1.5 cm (i.e., 1.5 cm from the bottom of the sample).

For each polymer dosage used and the Leda clay, 3 replicate columns were used for performing the fall cone test periodically. For keeping the boundary condition unchanged over time, the bleed water accumulated on the surface of the samples in each column was removed carefully using a syringe with a fine needle before performing the fall cone test, and the same bleed water was restored after the test. For the verification of the success of these protocols, the mass of each column was recorded before and after the removal of bleed water, as well as after the measurement of undrained shear strength, and finally, after the restoration of bleed water. The cone used had a weight of 40 g and an angle of 30^0. A cone factor (k) of 1.03, based on this cone angle and the cone surface roughness (Hansbo, 1957; Koumoto and Houlsby, 2001) was employed. The undrained shear strength was determined from the penetration value using the following empirical relationship proposed by Hansbo (1957):

$$s_u = \frac{kmg}{d^2} \tag{1}$$

where s_u = undrained shear strength (in kPa); k = cone factor, an empirical constant that depends on the apex angle of the cone, the cone surface roughness, and whether the sample is undisturbed or remoulded; m = mass of the cone; g = gravitational acceleration = 9.81 m/s^2; and d = depth of cone penetration (in mm).

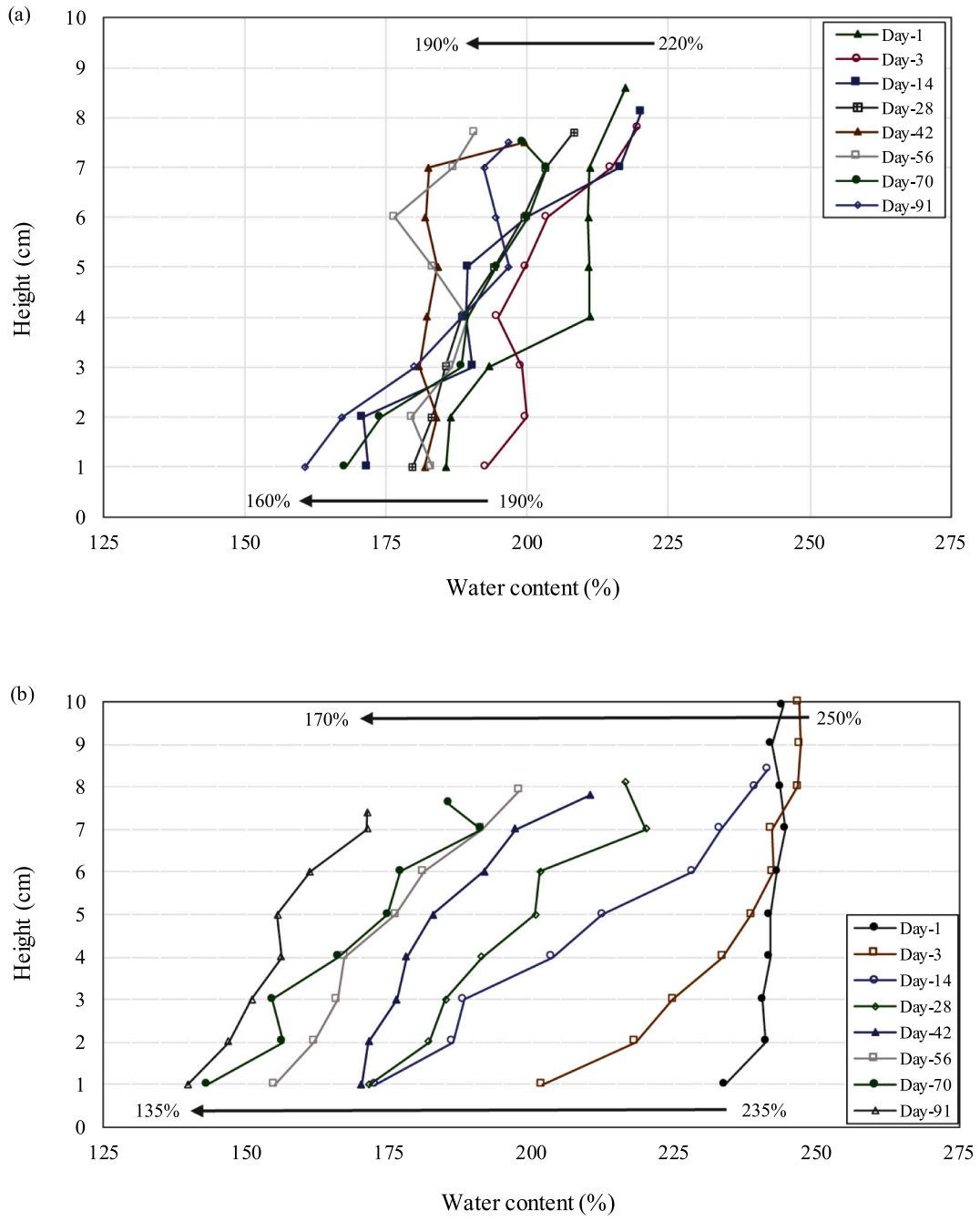

Fig. 6. Water content depth profiles in (a) 1000-ppm samples and (b) 600-ppm samples. The LL of these samples is ~80% (1000-ppm) and 70% (600-ppm).

Oedometer tests were performed on the aged samples collected from the bottommost layer of the replicate columns at specific time intervals (14-day, 35-day, 63-day, and 91-day). Columns specially customized for subsampling were used for the above samples. Before polymer-amended tailings were poured into those columns, the bottommost 2 cm part of the column (with the sealed base) was carefully cut and then again glued to the remaining part of the column using silicone adhesive sealant. To confirm that there would be no leakage, a strong waterproof tape was applied in multiple layers around the joints on the columns. Just a few moments before starting the consolidation test, the tape was carefully peeled off, and the 2-cm bottommost part of the column was removed using a wire saw without causing much disturbance to the sample. An oedometer ring (1.9 cm high) was carefully and very slowly pushed into the sample inside the 2 cm part of the column. Then, the ring with the sample inside was carefully removed and placed onto a porous stone. Immediately, the sample was mounted in the consolidation cell and the loading unit. The whole procedure took less than a minute and maximum caution was practiced to minimize sample disturbance; however, it was not possible to completely avoid sample disturbance when the oedometer ring is pushed into the sample as the procedure involves some penetration of the specimen by the cutting edge of the ring. Although minimal, such disturbance could possibly lead to a reduction in the yield stress and the pre-yield stiffness, as noted by Hight et al. (1992).

Since the flocculated FFT samples had high initial moisture contents, low consolidation stresses were used during oedometer tests to prevent the sample from squeezing through the space between the confining ring and the porous disk. The individual dead weights used were lower than conventional weights, and the load was confirmed using a load cell. The additional total stress due to self-weight was small due to the thinness of

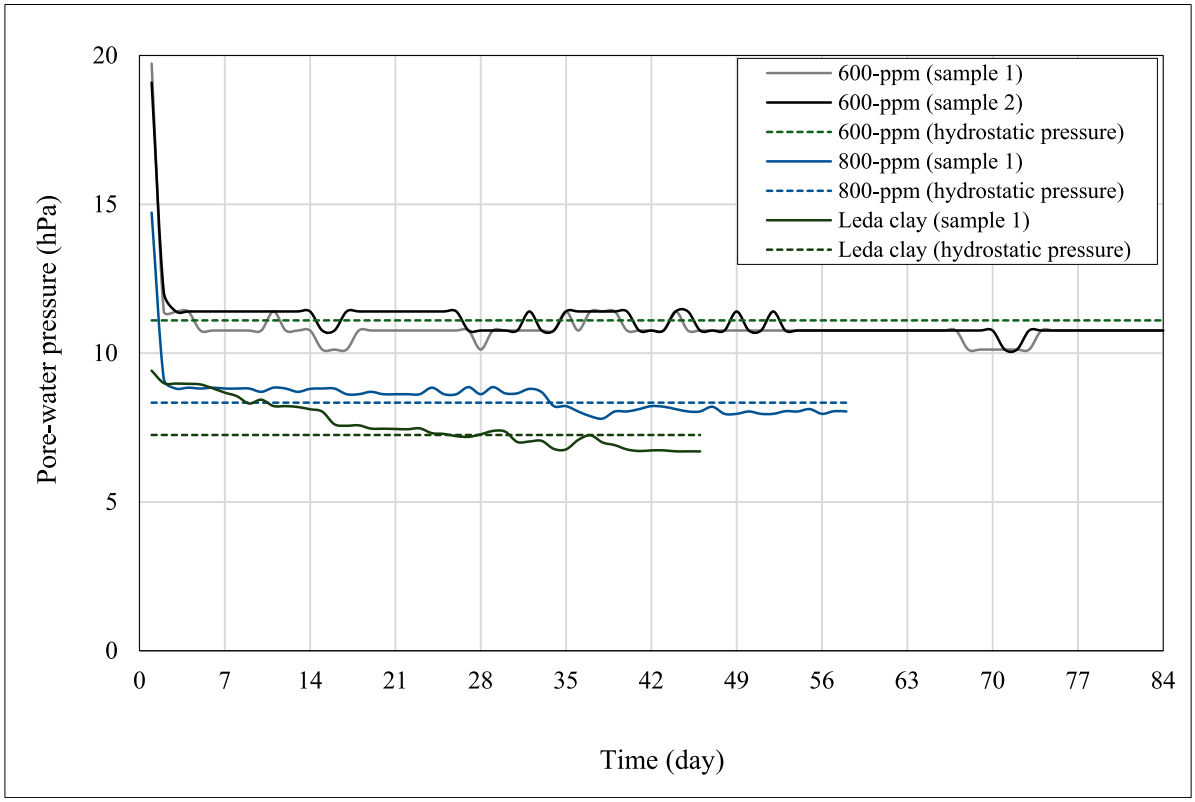

Fig. 7. Pore-water pressure measured (at an elevation of 1.5 cm) from the 600-ppm samples, 800-ppm sample, and Leda clay sample.

the sample (< 0.02 m), <0.4 kPa. The loading steps used for the samples tested in the incremental load oedometer were 1.7 kPa, 3.4 kPa, 6.9 kPa, 13.7 kPa, 27.5 kPa, 54.9 kPa, 109.9 kPa, 219.8 kPa, 439.6 kPa, and 879.3 kPa. Similar loading steps were used for soft reconstituted clays/dredged clays (Zeng et al., 2016; Shahriar et al., 2018). The Casagrande (1936) was used for determining the pre-consolidation pressure from the compressibility curves based on the point of greatest curvature.

4. Results

4.1. Dewatering and settlement behaviour

The average water contents of both the Leda clay and flocculated FFT (fFFT) samples are shown in Fig. 3 (please note that two y-axes are used to display these results). The average gravimetric water content in the Leda clay samples declined to 82% from 99% over 91 days (Fig. 3). The water content gradually declined till day 35; however, the change in water content became minimal afterwards, and the volume of the bleed water that accumulated on the surface of the solids with time was also nominal. Similarly, after day-35, the clay samples showed almost no settlement. The water content profiles with depth showed that the profiles were relatively uniform with depth (Fig. 4). This initial 35-day period corresponds to the typical period of self-weight consolidation (Fig. 7).

Interestingly, in the later stages of the experiment (> 63 days) the proportion of "free" to "absorbed" water changed, as defined by the proportion of water removed by 24 h drying to 105 °C versus 200 °C (Nutting, 1943; Mielenz et al., 1953). As indicated by Fig. 5, slight changes in this proportion seemed to indicate the possible transition of free water to interlayer or absorbed water, which might be an additional mechanism for ageing in the Leda clay.

All the fFFT samples showed persistent bleed water accumulation in all columns. Although the fFFT samples that were dosed with higher polymer dosages such as 1000-ppm samples had the highest initial water contents, they also showed a sharp decline in their water contents at the early stage of the experiment. For the first one to three days, the rate of change in water content was very high in 1000-ppm and 800-ppm samples, as well as in Leda clay samples; however, the rate of change in water content eventually declined in those samples. After 28 days, 1000-ppm samples showed very little change in their water contents (Fig. 3). By contrast, the fFFT samples that were dosed with relatively lower polymer dosages such as 600-ppm samples showed a relatively high rate of dewatering (down to $w = 155\%$). However, the self-weight consolidation period was confined to <1–2 weeks for all the fFFT samples (Fig. 7). Therefore, creep appears to be relatively larger in the lower dosage samples, while relatively small in the 1000-ppm sample. This behaviour is evident in the depth profiles of water contents (Fig. 6), which show the development of a varying water content profile with depth consistent with consolidation during early dewatering, following by dewatering which occurs more uniformly with depth.

4.2. Undrained shear strength

The strength buildup in the flocculated FFT samples varied depending on the polymer dosage. 1000-ppm and 800-ppm samples developed the highest strengths, whereas 600-ppm samples developed a nominal strength (Fig. 8). Initially, 1000-ppm and 800-ppm samples developed shear strength at a minimal rate; however, once the rate of initial dewatering or consolidation decreased, the strength started to increase substantially. Hence, the change in void ratio or density was thought to be an insignificant factor in the strength gain in 800-ppm and 1000-ppm samples.

Examples of pore-water pressure data are given in Fig. 7. Similar data is also reported in the published thesis of Salam (2020) (publicly available). This data implies that self-weight consolidation finishes within 1 week in the fFFT samples (i.e., 600-ppm and 800-ppm), and within 25–35 days in the natural clay (i.e., Leda clay), i.e., well before the onset of the second increase in strength as shown in Fig. 8.

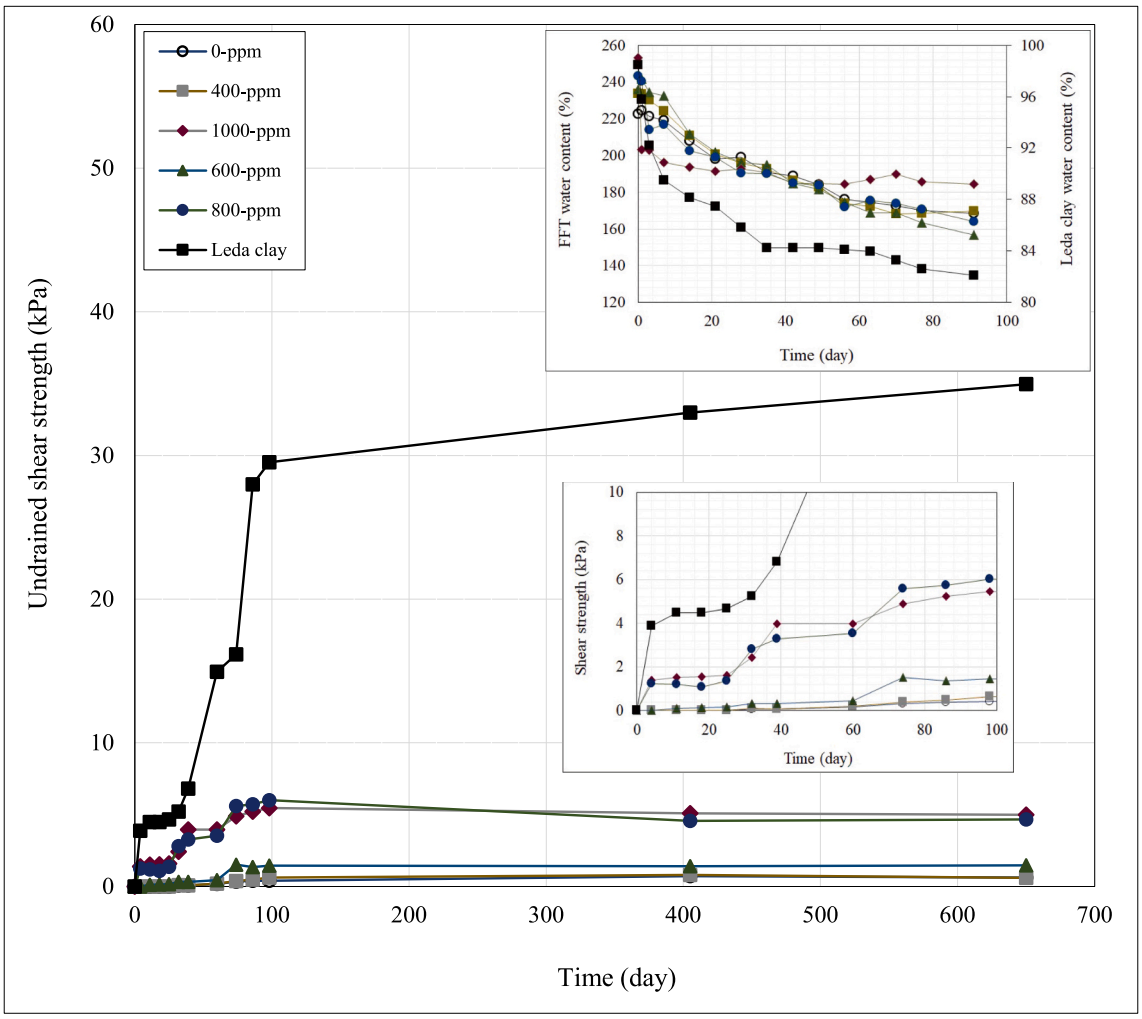

Fig. 8. Undrained shear strength development in the Leda clay and flocculated FFT samples, in normal scales, with an insert at low strengths and times <100 Days.

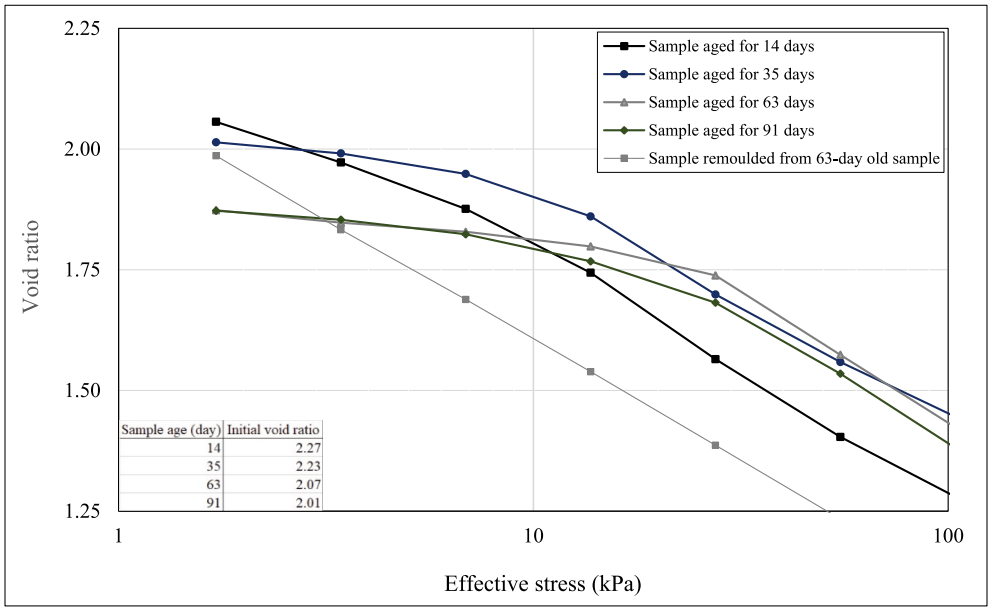

Fig. 9. Compressibility behaviour of the Leda clay samples that aged for 14, 35, 63, and 91 days and of the remoulded sample which was prepared (at the same water content) from the Leda clay that aged for 63 days.

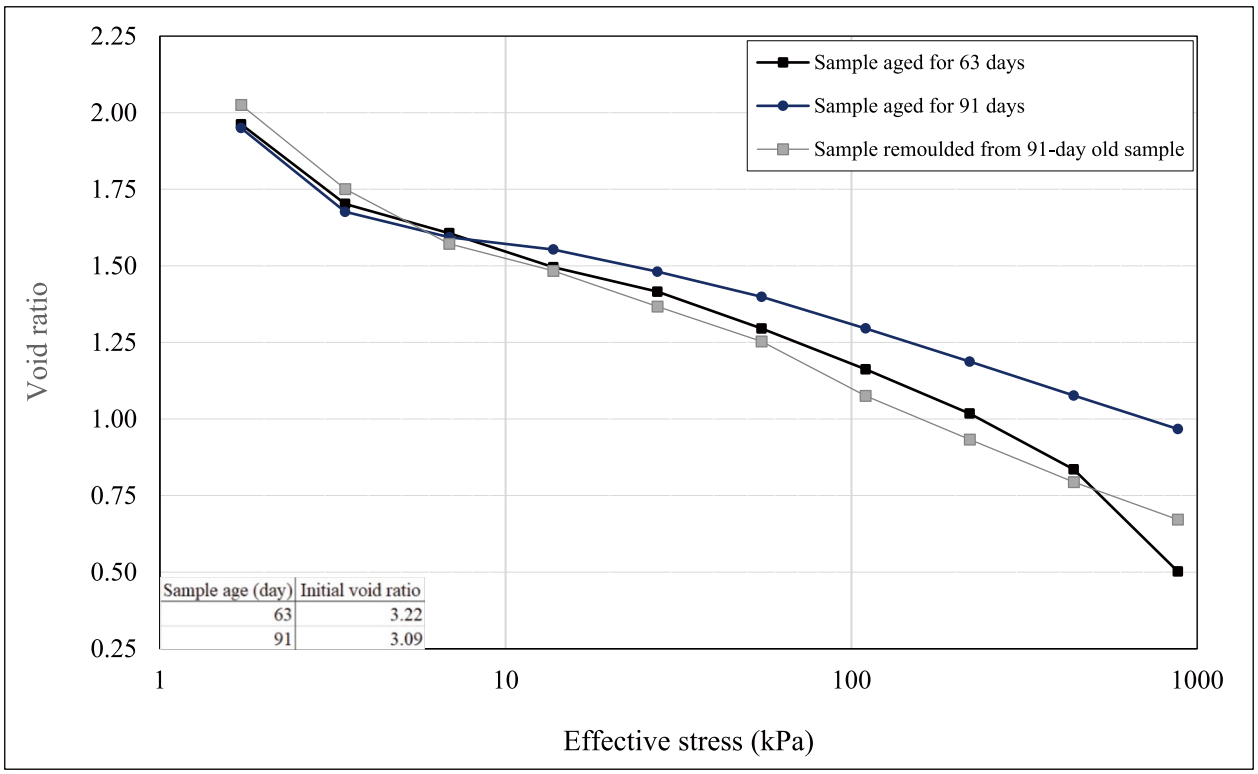

Fig. 10. Compressibility behaviour of the 600-ppm samples that aged for 63 and 91 days and of the remoulded sample that was prepared (at the same water content) from the 600-ppm sample that aged for 91 days.

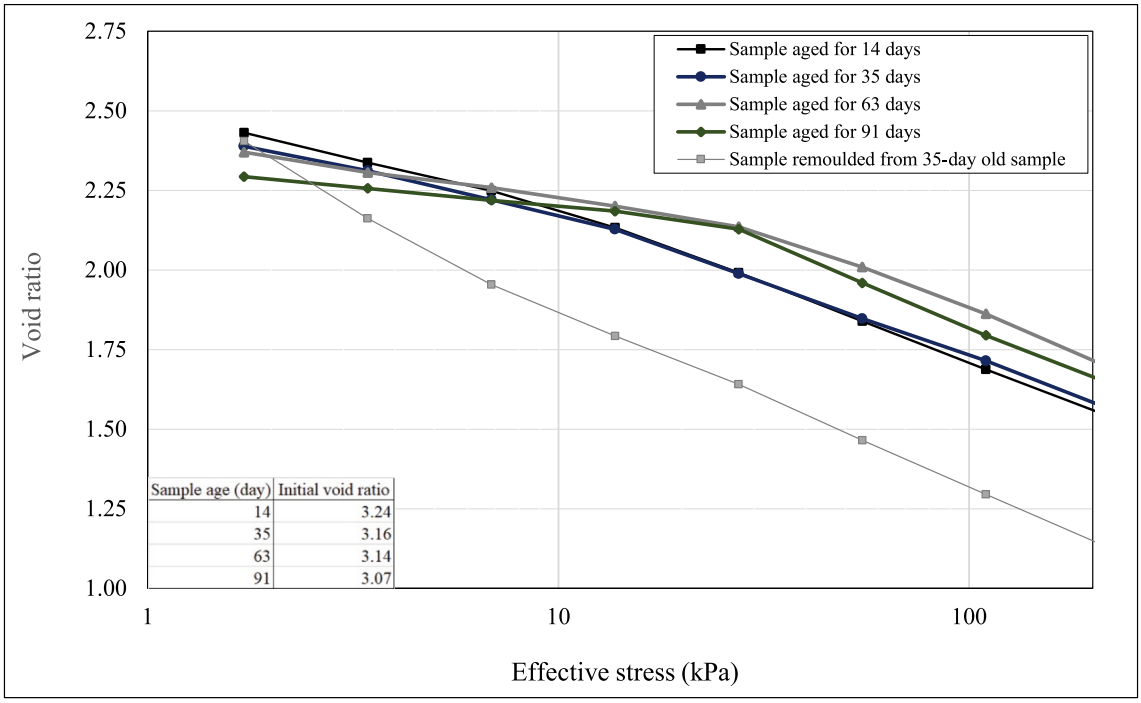

Fig. 11. Compressibility behaviour of the 1000-ppm samples that aged for 14, 35, 63, and 91 days and of the remoulded sample that was prepared (at the same water content) from the 1000-ppm sample that aged for 35 days.

When the undrained shear strength development in Leda clay samples was compared to the strength development in flocculated FFT samples, both Leda clay and FFT samples developed low shear strengths at the beginning though the strengths developed in clay samples were relatively higher. The strengths remained at the almost same level up to day-25. But, once the samples started to reach an equilibrium in terms of water content, the strengths began to increase. The magnitude of strength development in Leda clay samples was significantly higher than that in the flocculated FFT samples; among those, the higher polymer dosage samples developed relatively higher strengths.

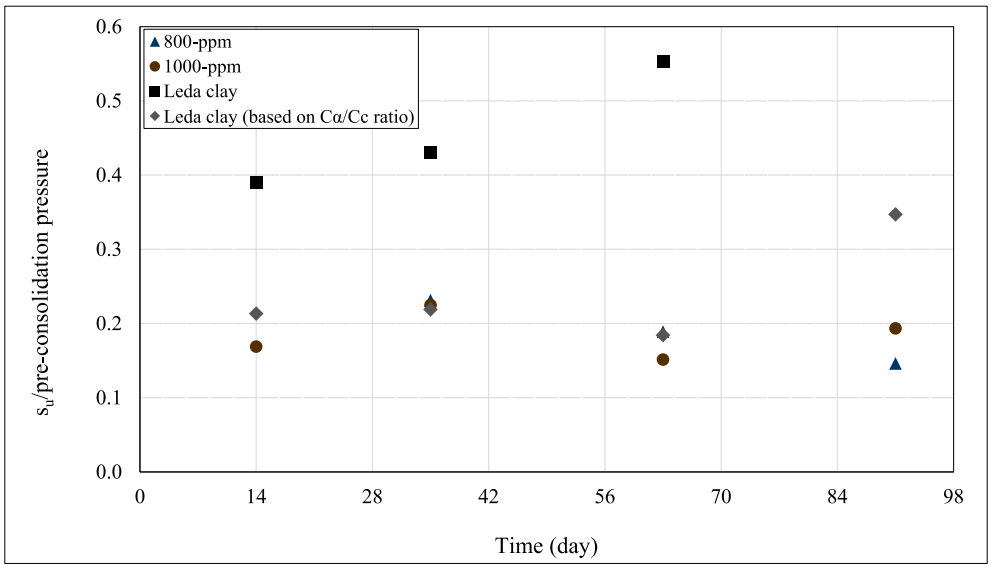

Fig. 12. The ratio of strength to pre-consolidation pressure (800-ppm, 1000-ppm, Leda clay).

4.3. Compressibility and apparent pre-consolidation pressure

Oedometer tests were performed on the Leda clay and flocculated FFT samples at specific times. In addition to the samples that were re-sedimented by the self-weight of the material in the 10 cm columns over time, oedometer tests were also performed on the remoulded samples prepared from those samples at the same water contents. The compressibility curves obtained from the 14, 35, 63, and 91-day old Leda clay samples showed a change in compressibility behaviour over time (Fig. 9) and exhibited an increase in pre-consolidation pressure (measured using the Casagrande method), as well as a decrease in the slope of the compressibility curve before the pre-consolidation pressure, while the slope of the compressibility curves after the pre-consolidation pressure was relatively similar (and was similar to the remoulded curve). The initial void ratios in Fig. 9 correspond to the void ratios estimated from the samples based on the measurement of their initial and final sample heights, before and after consolidation testing respectively. Since the flocculated FFT samples had high initial moisture contents when loaded, there was a substantial reduction in void ratio even during the first loading step.

When oedometer tests were performed on the flocculated FFT samples, the 0-ppm and 400-ppm samples remained too soft for the entire experiment for conducting consolidation tests, whereas the 600-ppm samples were able to sustain the consolidation loads after 35 days. 63-day old 600-ppm sample showed some signs of developing stiffness. 91-day old 600-ppm sample showed relatively greater stiffness, but still, there was no distinct pre-consolidation pressure development (Fig. 10). By contrast, the samples dosed with higher dosages such as 1000-ppm showed considerably lower compressibility and developed distinct pre-consolidation pressures starting from 14 days (Fig. 11).

In addition to the Casagrande method, the 'work' method proposed by Becker et al. (1987) was used to estimate the pre-consolidation pressures for the Leda clay and 1000-ppm samples. When compared to the Casagrande method, the method proposed by Becker et al. (1987) was found to estimate pre-consolidation stresses that are about 5– 10% lower than the Casagrande values.

5. Discussion

5.1. Correlations of shear strength and pre-consolidation pressure, and comparison of compression curves with the intrinsic and sedimentation lines proposed by Burland (1990)

The undrained shear strengths of 1000-ppm and 800-ppm samples were normalized by the pre-consolidation pressures. It was found that the ratio between undrained shear strength and pre-consolidation pressure of 0.18 was more or less constant for the different ageing times (with Pearson's correlation coefficient of $R = -1.00$ for 800-ppm and $R = -0.03$ for 1000-ppm samples) (Fig. 12):

$$\frac{s_u}{\sigma_p'} = 0.18 \tag{2}$$

where s_u = undrained shear strength; and σ_p' = pre-consolidation pressure.

Given the accuracy of the data, the tailings seem to conform to the well-established ratio of shear strength to pre-consolidation pressure proposed by Mesri (1975) of 0.22. Although the value of 0.22 proposed by Mesri (1975) was for the undrained shear strength mobilized at failure under embankments, Mesri (1989) obtained the same value (~ 0.22) for the undrained shear strength mobilized in soft clays from laboratory shear tests where the effects of anisotropy and strain rate were considered. However, while the Leda clay samples also showed a strong correlation between shear strength and pre-consolidation pressure (Fig. 12), the ratio is much higher (~ 0.5) based on Casagrande pre-consolidation pressures, but similar (0.24) when using the C_α/C_c ratio. Calculating the C_α/C_c ratio is another way to present the compressibility data, as shown in Figs. A1 and A2 in the Appendix for the selected 1000-ppm fFFT and Leda clay samples. The pre-consolidation pressure generally falls just below the peak C_α/C_c ratio, as shown by Graham et al. (1983). In other studies of Leda clay, shear strength element tests such as simple shear give ratios of shear strength very close to 0.22 (Theenathayarl, 2015). Field studies on Leda clay deposits report in-situ ratios in a similar range, for example, 0.26 is reported in a recent summary of field studies at the Gloucester Test Site (Mayne et al., 2019), while the ratio between field vane tests and pre-consolidation pressures reported for several sites in Leda clay by Quigley et al. (1983) indeed reaches as high as 0.5. These higher ratios seem to reflect a more gradual transition between over-consolidated and normally consolidated states (e.g., Boone, 2010).

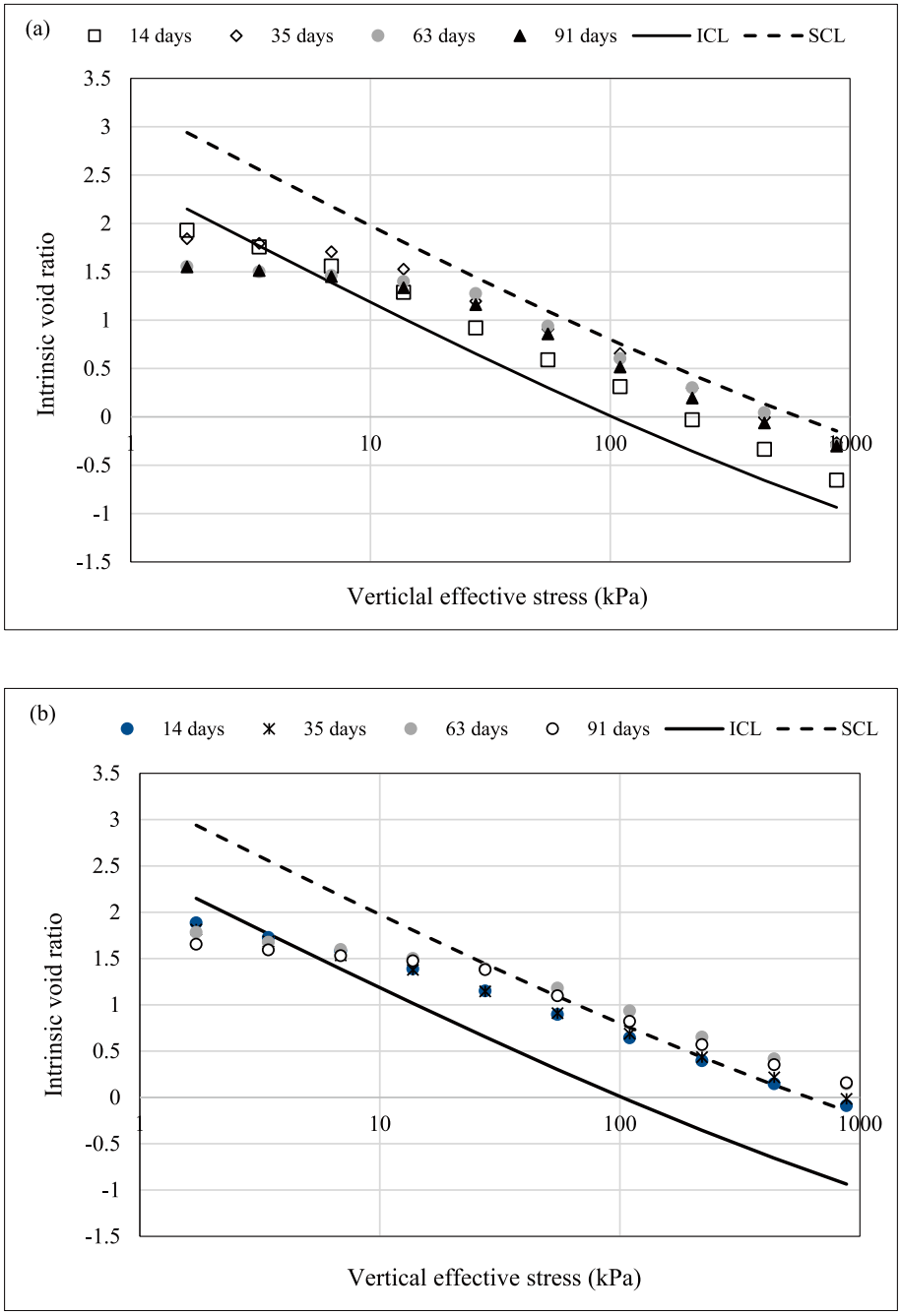

Fig. 13. (a) Leda clay and (b) 1000-ppm fFFT compression curves plotted as intrinsic void ratios and compared with ICL and SCL of Burland (1990).

It is also interesting to compare the change in compressibility curves in terms of the intrinsic compression lines (ICL) and sedimentary compression lines (SCL) proposed by Burland (1990). Fig. 13 compares the ICL and SCL to the Leda clay and 1000-ppm fFFT compression curves, transformed to intrinsic void ratio using the void ratio at 100 and 1000 kPa measured during the oedometer test of a remoulded sample. The compression curves of the aged Leda clay samples approach the SCL and indeed carry about 5 times the load at a given effective stress compared to the remoulded sample, which was shown by Burland to be the case for a wide range of natural clay soils. It appears, for the Leda clay, clay approaches its "natural behaviour" within about 100 days. For the tailings, the difference between the remoulded curve and the oldest compression curve is closer to 10, and the most aged sample plots somewhat to the right of the SCL. This likely reflects the action of the polymer, where some of the flocs created by the polymer are irreversibly destroyed during remoulded. Consequently, the remoulded compression curve for the tailings will exhibit a greater difference from the compression curve at maximum ageing, compared to the Leda clay.

5.2. *Linking degree of ageing to the EES*

Similar to the dual-mode strength development behaviour shown by ideal clay suspensions in ageing experiments, which was characterized by a strength development in the sample in two distinct phases (as shown in Fig. 1), in the present study, when looking at the individual undrained shear strength curves, there appears to be such an "early equilibrium state" (EES) in all the tests (Fig. 14). However, this early equilibrium state may also require the end of any initial sedimentation and consolidation. The "early equilibrium state" (EES) strength can be defined by two conditions i) the shear strength closest to the change in

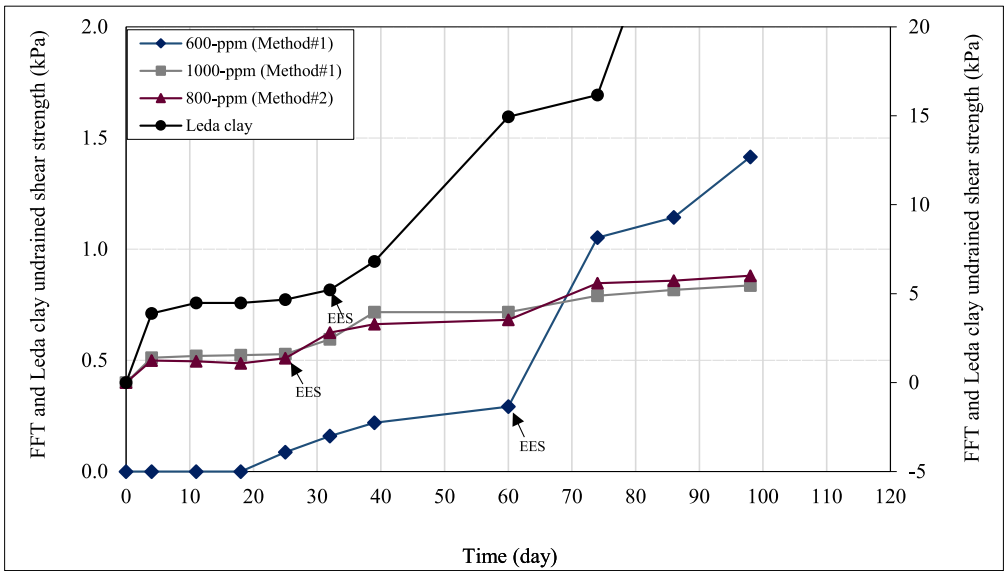

Fig. 14. The determination of Early Equilibrium State (EES) for Leda clay, 600-ppm, 800-ppm and 1000-ppm fFFT samples (determined from the shear strength closest to the change in curvature, in other words, when the decreasing slope changes to an increasing slope).

curvature, in other words when the decreasing slope changes to an increasing slope and ii) occurring after the end of primary consolidation, as indicated by pore-water pressure readings in Fig. 7. In all the data sets where ageing is significant, the EES is quite unambiguous using these criteria.

The correlation between the EES and the magnitude of the ageing effect is interesting and potentially quite useful, if the EES is a reliable and robust predictor of future ageing behaviour. Before examining the potential of EES as a predictor of future ageing, other ageing data sets from the literature are introduced. Figs. 15(a) and (b) include data taken from Fig. 5 in Skempton and Northey (1952) and Fig. 3 in Zhang et al. (2017). Data from Skempton and Northey (1952) is given in terms of percent strength regain, defined by those authors as "(Actual strength – remoulded strength)/remoulded strength."

While some data sets have an EES that can be obtained using the criteria in the previous section, such as the "Detroit" or "London" data set, others do not. The shape of all these data sets, however, seems to be well described by an S-shaped curve in log-time. Therefore, the logistic function is proposed as a descriptive (though not predictive) equation for these data sets.

$$f(x) = \frac{L}{1 + e^{-k(x-x_0)}} \tag{3}$$

The parameters of the equation are L, k, and x_0. L is the maximum value of strength corresponding to the second plateau of the ageing data. The time x_0 corresponds to the time, where strength $= L/2$. These two values can be determined unambiguously from all the data sets. The last parameter, k is the average slope and can be determined by any curve-fitting exercise. As an example, "L" for the London data set would be the value of the last point or 85%. The time corresponding to $L/2$ (42.5%), by interpolating the two closest data points, gives 40 days. To get the best fit, as selected by minimizing the mean average error (MAE) associated with all data points, gives a k value of 0.04. Fits of the logistic function with the literature data are presented in Fig. 16.

This function will have two asymptotic values at $t >$ infinity, and at $t > 0$, the former L the maximum value, and latter the EES value, which would be $EES = \frac{L}{1+exp\ (kx_0)}$, and is therefore explicitly given by the fit of the logistic curve.

Once the EES is obtained, the strength curves of the literature data and the data from this paper on the fFFT and the Leda clay are summarized in Fig. 17, in which the strength is normalized with each EES. Except for the Shellhaven data set, the ageing curves are quite similar. It is interesting to recall again the observation of Burland (1990) regarding the difference in load supported by natural clays and by their reconstituted equivalent, being about a factor of 5 for a wide range of clays. The EES is effectively the reconstituted strength, that is remoulding plus consolidation. The normalized strength results in Fig. 17 seem congruent with the observation of Burland (1990) and should be investigated further.

5.3. The practical implication for polymer dosage and the ageing effect

Although optimal flocculation as judged by short-term dewatering is given by the 1000-ppm and 800-ppm samples, an increase in strength/stiffness over time would reduce the magnitude of consolidation in real tailings deposits, which may be quite deep (> 50 m). By contrast, the samples dosed with relatively lower polymer dosages such as 600-ppm would be substantially more compressible in deep deposits. A simple comparative analysis of the final density distribution of a hypothetical 50 m deposit using the aged compressibility curves for 600-ppm and 1000-ppm are shown in Fig. 18, based on fitted power functions to the compressibility curve, and obtaining from the theoretical steady-state profile at complete dissipation of excess pore-water pressure. The following equation (Qi and Simms, 2019) is used for determining the final height of the deposit:

$$\xi = \frac{1}{\Upsilon}\left[\left(\frac{e}{a}\right)^{\frac{1}{b}}\left(\frac{e}{b+1}+1\right) - c\left(\frac{ac^b}{b+1}+1\right)\right] \tag{4}$$

where ξ is the depth below the tailings-water interface, $\Upsilon = (G_s - 1)$ x unit weight of water, e is the void ratio, and a, b, and c are parameters of the function used to fit the compressibility data, $e = a\acute{\sigma}_v{}^b + c$ (where $\acute{\sigma}_v{}^b$ is the effective stress).

Predicted final density profiles are shown for initial void ratios of 5 and initial heights of deposit of 50 m. The substantial difference in the density profiles has clear implications for the stability of deposits for purposes of terrestrial reclamation. The void ratios of the 1000-ppm sample are all well above the liquid limit at LI (Liquidity index) > 1.5, whereas most of the 600-ppm deposit is below the liquid limit ($e = 1.5$). Clearly, these deposits would have substantially different strength profiles, the 1000-ppm sample being considerably more difficult to reclaim.

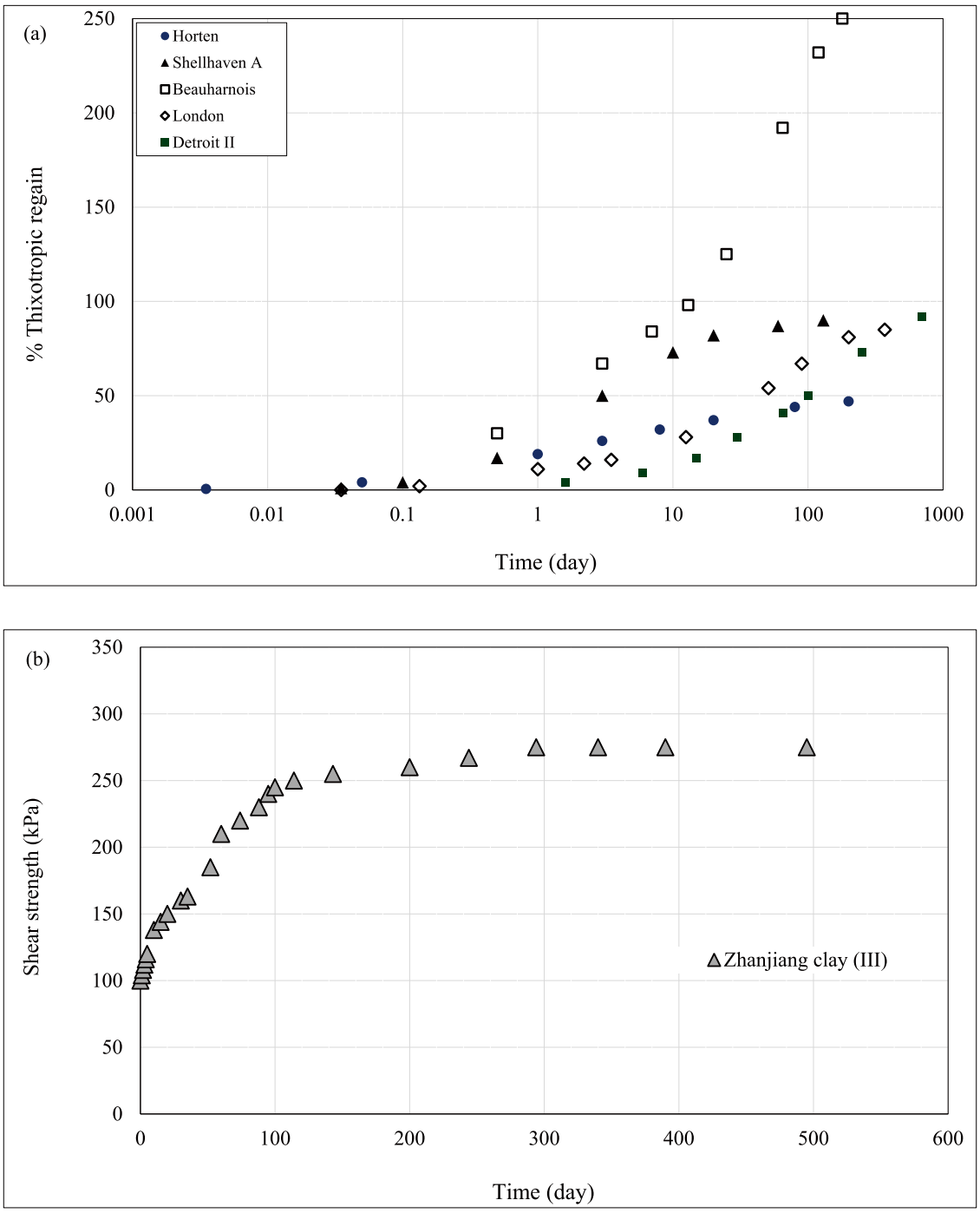

Fig. 15. (a) Fig. 5 from Skempton and Northey, 1952 (adapted); (b) Fig. 3 from Zhang et al., 2017 (adapted).

6. Conclusions

Short (10 cm tall) sealed columns of polymer-amended tailings and a natural clay prepared at twice its LL were left to age for up to 650 days. The main findings of the research presented in this paper are:

1. Both samples of polymer-amended clayey tailings, and a natural clay, showed qualitatively similar increases in shear strength and pre-consolidation pressures, as measured using a fall cone and oedometer tests.
2. The shear strength to pre-consolidation pressure ratio for the tailings (0.18) was very close to the commonly used ratio of Mesri (1975) (0.22), while the ratio of fall cone shear strength to Casagrande pre-consolidation pressure of Leda clay was higher (0.5).
3. Samples showing a large amount of ageing were associated with a relatively high decrease in water content early in the test (in the first few days), but also with a relatively little decrease in water content thereafter, especially during the period congruent with ageing.
4. The magnitude of the ageing phenomenon is sufficient to cause substantial differences in consolidation and settlement in deep deposits of soft tailings, with important implications as to the feasibility of reclamation of those deposits.
5. The ageing behaviour could be defined by two separate modes. The early mode is called the Early Equilibrium State, which seems associated with the end of primary consolidation. In the second mode, the

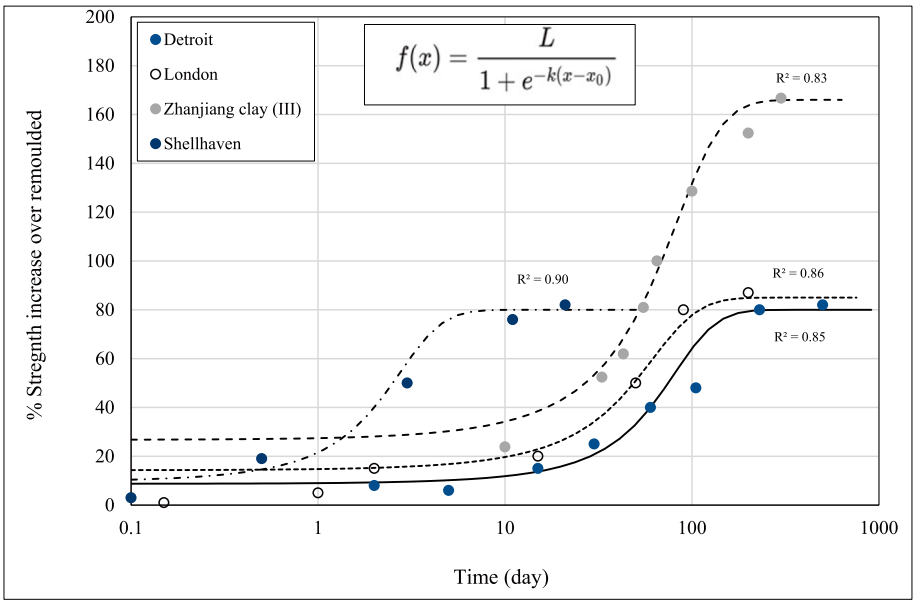

Fig. 16. Logistic function fitted to (a) Detroit, (b) London, (c) Shellhaven, and (d) Zhanjiang (III) clays to determine the EES.

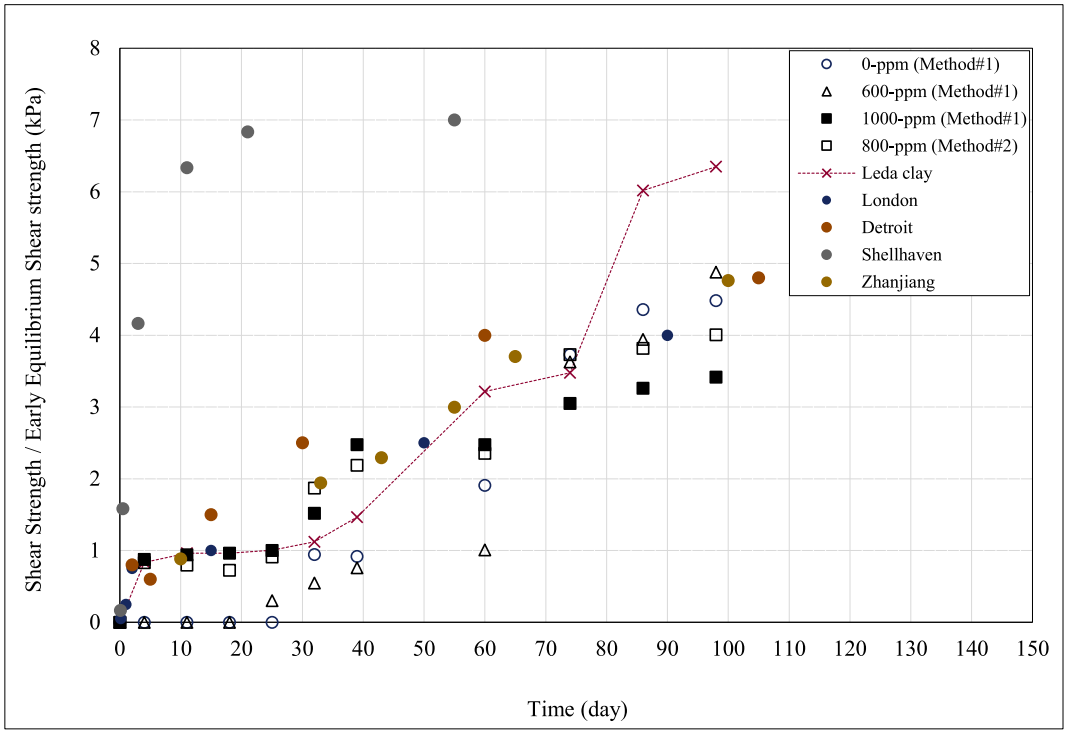

Fig. 17. Increase in absolute shear strength above the remoulded state, relative to the Early Equilibrium State (EES).

strength increases up to some maximum, which in our samples occurred at ~100 days. The second mode occurs after the completion of self-weight consolidation and occurs simultaneously with a relatively small amount of dewatering.

6. Some other soils from the literature appear to follow the dual-mode behaviour and show similar ageing behaviour when their strength is normalized to the EES.

In terms of general behaviour of soft soils, the most potentially important finding is the EES and its potential application as a predictor for long-term ageing. This hypothesis should be explored using a wider range of soils and tailings, under different loading conditions.

CRediT authorship contribution statement

Muhammad A. Salam: Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization. **Paul H. Simms:** Conceptualization, Methodology, Validation, Formal analysis, Resources, Writing – review & editing, Visualization, Supervision, Project administration, Funding acquisition. **Banu Örmeci:** Validation, Writing – review & editing, Visualization, Supervision.

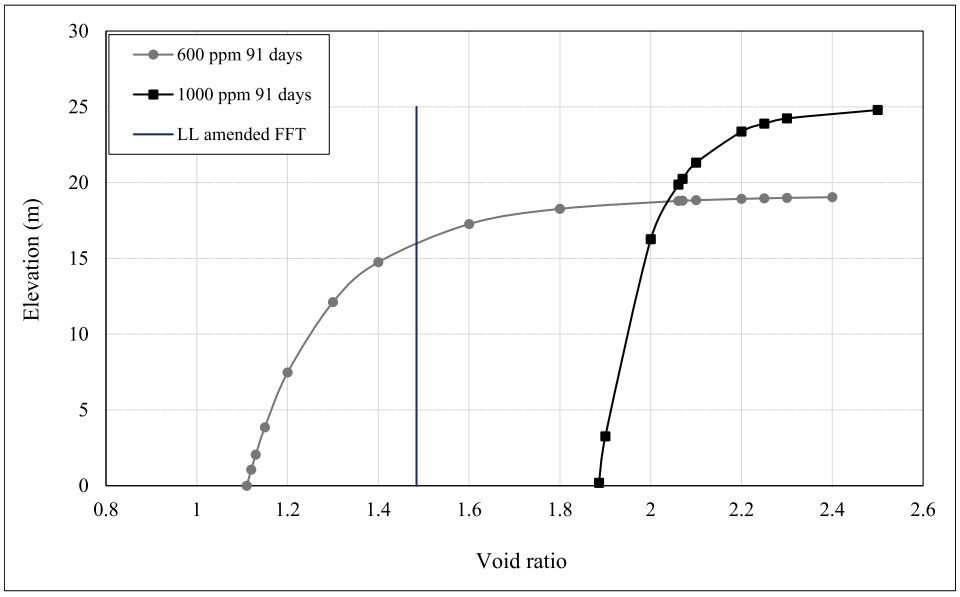

Fig. 18. Final void ratio depth profiles for deposits with initial void ratios of 5 and initial heights of 50 m, based on the aged compressibility functions of either 600-ppm or 1000-ppm samples, and using Eq. 4.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.enggeo.2023.107088.

References

Abou, B., Bonn, D., Meunier, J., 2001. Aging dynamics in a colloidal glass. Phys. Rev. E 64 (2), 021510.
Alam, M.K., Shahriar, A.R., Islam, M.S., Islam, N., Abedin, M.Z., 2021. Experimental investigation on the strength and deformation aspects of thixotropic aging in reconstituted clays. Geotech. Geol. Eng. 39 (3), 2471–2486.
Anik, K.A., 2019. Shear Strength and Compressibility Characteristics of Reconstituted Clays.
Au, P.I., Leong, Y.K., 2015. Surface chemistry and rheology of Laponite dispersions—Zeta potential, yield stress, ageing, fractal dimension and pyrophosphate. Appl. Clay Sci. 107, 36–45.
Banas, L.C., 1991. Thixotropic Behaviour of Oil Sands Tailings Sludge (Master's Thesis). The University of Alberta, Edmonton, Alberta.
Becker, D.E., Crooks, J.H.A., Been, K., Jefferies, M.G., 1987. Work as a criterion for determining in situ and yield stresses in clays. Can. Geotech. J. 24 (4), 549–564.
Bonacci, F., Chateau, X., Furst, E.M., Fusier, J., Goyon, J., Lemaître, A., 2020. Contact and macroscopic ageing in colloidal suspensions. Nat. Mater. 1–6.
Boone, S.J., 2010. A critical reappraisal of "preconsolidation pressure" interpretations using the oedometer test. Can. Geotech. J. 47 (3), 281–296.
Burland, J.B., 1990. On the compressibility and shear strength of natural clays. Géotechnique 40 (3), 329–378.
Casagrande, A., 1936. The determination of pre-consolidation load and it's practical significance. In: Proc. Int. Conf. Soil Mech. Found. Eng. Cambridge, Mass., Vol. 3, p. 60.
Crawford, C.B., 1968. Quick clays of eastern Canada. Eng. Geol. 2 (4), 239–265.
Goh, R., Leong, Y.K., Lehane, B., 2011. Bentonite slurries—zeta potential, yield stress, adsorbed additive and time-dependent behaviour. Rheol. Acta 50 (1), 29–38.
Graham, J., Crooks, J.H.A., Bell, A.L., 1983. Time effects on the stress-strain behaviour of natural soft clays. Géotechnique 33 (3), 327–340.
Hansbo, S., 1957. New Approach to the Determination of the Shear Strength of Clay by the Fall-Cone Test.
Hight, D.W., Böese, R., Butcher, A.P., Clayton, C.R.I., Smith, P.R., 1992. Disturbance of the Bothkennar clay prior to laboratory testing. Géotechnique 42 (2), 199–217.
Hinchberger, S.D., Rowe, R.K., 2005. Evaluation of the predictive ability of two elastic-viscoplastic constitutive models. Can. Geotech. J. 42 (6), 1675–1694.
Jeeravipoolvarn, S., 2005. Compression Behaviour of Thixotropic Oil Sands Tailings (Master's Thesis). The University of Alberta, Edmonton, Alberta.
Jeeravipoolvarn, S., Scott, J.D., Chalaturnyk, R.J., 2009. 10 m standpipe tests on oil sands tailings: long-term experimental results and prediction. Can. Geotech. J. 46 (8), 875–888.
Koumoto, T., Houlsby, G.T., 2001. Theory and practice of the fall cone test. Géotechnique 51 (8), 701–712.
Lerouеil, S., Vaughan, P.R., 1990. The general and congruent effects of structure in natural soils and weak rocks. Géotechnique 40 (3), 467–488.
Lerouеil, S., Perret, D., Locat, J., 1996. Strain rate and structuring effects on the compressibility of a young clay. In: Measuring and Modeling Time Dependent Soil Behavior. ASCE, pp. 137–150.
Liu, J., Afroz, M., Ahmad, A., 2021. Experimental investigation of the impact of salinity on Champlain Sea clay. Mar. Georesour. Geotechnol. 39 (4), 494–504.
Liu, M.D., Carter, J.P., 1999. Virgin compression of structured soils. Géotechnique 49 (1), 43–57.
Locat, J., Lefebvre, G., 1986. The origin of structuration of the Grande-Baleine marine sediments, Québec, Canada. Q. J. Eng. Geol. Hydrogeol. 19 (4), 365–374.
Mayne, P.W., Cargill, E., Miller, B., 2019. Geotechnical characteristics of sensitive Leda clay at Canada test site in Gloucester, Ontario. AIMS Geosci. 5, 390–411.
Mesri, G., 1975. New design procedure for stability of soft clays. J. Geotech. Geoenviron. 101 (Discussion).
Mesri, G., 1989. A reevaluation of using laboratory shear tests. Can. Geotech. J. 26 (1), 162–164.
Mewis, J., Wagner, N.J., 2012. Colloidal Suspension Rheology. Cambridge University Press.
Mielenz, R.C., Schieltz, N.C., King, M.E., 1953. Thermogravimetric analysis of clay and clay-like minerals. Clay Clay Miner. 2 (1), 285–314.
Miller, W.G., 2010. Comparison of Geoenvironmental Properties of Caustic and Noncaustic Oil Sand Fine Tailings (Ph.D. Thesis,. the University of Alberta.
Mitchell, J.K., 1960. Fundamental aspects of thixotropy in soils. J. Soil Mech. Found. Div. 86 (3), 19–52.
Mizani, S., Simms, P., Wilson, W., 2017. Rheology for deposition control of polymer-amended oil sands tailings. Rheol. Acta 56 (7–8), 623–634.
Monsif, M.Y., Liu, J., Gurpersaud, N., 2021. Microstructural analyses of cement-based binders in stabilizing Champlain Sea clay. Geotech. Geol. Eng. 1–19.
Nutting, P.G., 1943. Some Standard Thermal Dehydration Curves of Minerals. US Government Printing Office.
Pujala, R.K., Bohidar, H.B., 2013. Slow dynamics, hydration and heterogeneity in Laponite dispersions. Soft Matter 9 (6), 2003–2010.
Qi, S., Simms, P., 2019. Robust methods to estimate large-strain consolidation parameters from column experiments. Can. Geotech. J. 57 (5), 683–705.

Quigley, R.M., Thompson, C.D., 1966. The fabric of anisotropically consolidated sensitive marine clay. Can. Geotech. J. 3 (2), 61–73.
Quigley, R.M., Gwyn, Q.H.J., White, O.L., Rowe, R.K., Haynes, J.E., Bohdanowicz, A., 1983. Leda clay from deep boreholes at Hawkesbury, Ontario. Part I: Geology and geotechnique. Can. Geotech. J. 20 (2), 288–298.
Ruzicka, B., Zulian, L., Ruocco, G., 2007. Ageing dynamics in Laponite dispersions at various salt concentrations. Philos. Mag. 87 (3–5), 449–458.
Salam, A.M., 2020. Effects of Polymers on Short-and Long-Term Dewatering of Oil Sands Tailings (Doctoral dissertation). Carleton University.
Salam, A.M., Simms, P.H., Örmeci, B., 2018a. Structuration in polymer amended oil sands fine tailings. In: Proceedings of GeoEdmonton 2018, 71st Canadian Geotechnical Conference. Edmonton, Alberta.
Salam, A.M., Simms, P.H., Örmeci, B., 2018b. Evidence of Creep & Structuration in Polymer Amended Oil Sands Tailings. In: Proceedings of the 6th International Oil Sands Tailings Conference. Edmonton, Alberta.
Seng, S., Tanaka, H., 2012. Properties of very soft clays: a study of thixotropic hardening and behavior under low consolidation pressure. Soils Found. 52 (2), 335–345.
Shahriar, A.R., Jadid, R., 2018. An experimental investigation on the effect of thixotropic aging on primary and secondary compression of reconstituted dredged clays. Appl. Clay Sci. 162, 524–533.
Shahriar, A.R., Abedin, M.Z., Jadid, R., 2018. Thixotropic aging and its effect on 1-D compression behavior of soft reconstituted clays. Appl. Clay Sci. 153, 217–227.
Skempton, A.W., Northey, R.D., 1952. The sensitivity of clays. Geotechnique 3 (1), 30–53.
Sorensen, K.K., 2006. Influence of Viscosity and Ageing on the Behaviour of clays (Doctoral dissertation,. University College London.
Suthaker, N.N., 1995. Geotechnics of Oil Sand Fine Tailings (Doctoral Dissertation). The University of Alberta, Edmonton, Canada.
Suthaker, N.N., Scott, J.D., 1997. Thixotropic strength measurement of oil sand fine tailings. Can. Geotech. J. 34 (6), 974–984.
Tanaka, H., Jabbari-Farouji, S., Meunier, J., Bonn, D., 2005. Kinetics of ergodic-to-nonergodic transitions in charged colloidal suspensions: Aging and gelation. Phys. Rev. E 71 (2), 021402.
Theenathayarl, T., 2015. Behaviour of Sensitive Leda Clay under Simple Shear Loading (Doctoral dissertation,. Carleton University.
Vermeer, P.A., Neher, H.P., 1999. A soft soil model that accounts for creep. In: Beyond 2000 in Computational Geotechnics, pp. 249–261.
Walker, L.K., Raymond, G.P., 1968. The prediction of consolidation rates in a cemented clay. Can. Geotech. J. 5 (4), 192–216.
Weeks, E.R., 2017. Introduction to the Colloidal Glass Transition.
Yin, J.H., Graham, J., 1994. Equivalent times and one-dimensional elastic viscoplastic modelling of time-dependent stress-strain behaviour of clays. Can. Geotech. J. 31 (1), 42–52.
Yong, R.N., Nagaraj, T.S., 1977. Investigation of fabric and compressibility of a sensitive clay. In: Proceedings of the International Symposium on Soft Clay, Asian Institute of Technology, pp. 327–333.
Zeng, L.L., Hong, Z.S., Cui, Y.J., 2016. Time-dependent compression behaviour of dredged clays at high water contents in China. Appl. Clay Sci. 123, 320–328.
Zhang, X.W., Kong, L.W., Yang, A.W., Sayem, H.M., 2017. Thixotropic mechanism of clay: a microstructural investigation. Soils Found. 57 (1), 23–35.